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Author(s):	J. L. Thompson, CIC-8 M. A. Guell and J. R. Hunt, UC-Berkeley
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### Subsurface Nobel Gas Transport at the Nevada Test Site

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Joseph L. Thompson\* Los Alamos National Laboratory

Margaret A. Guell and James R. Hunt University of California-Berkeley

#### Abstract

This is the final report of a three-year, Laboratory Directed Research and Development (LDRD) project at Los Alamos National Laboratory (LANL). The objective of our research was to explain the results of a groundwater pumping test done from 1975 to 1991 at the location of the nuclear test "Cambric" on the Nevada Test Site. The elution data from the pumped well indicated that krypton was delayed relative to tritium in the eluate and that less than half of the calculated Kr-85 source term was removed (though over 92% of the tritium was removed). We postulated an explanation for these observations and tested it with a mathematical model that simulated the movement of tritium and krypton at this site. The model showed that the hypothesis was consistent with the observed behavior; but the model was very sensitive to assumptions about initial radionuclide distributions and to hydrologic parameters.

#### **Background and Research Objectives**

The US Department of Energy complex has numerous sites of environmental contamination caused by weapons testing and previous waste disposal practices. The wastes of concern for human health and the ecosystem include volatile organic solvents, trace metals, and radionuclides. Because of the broad range of contaminant types, species exist in a variety of physicochemical states. While some species are associated predominantly with solid material in the subsurface, others exist primarily in the liquid form. The latter includes noble gases produced by weapons testing which partition between air and water. It is important to apply environmental transport models for these different classes of contaminants in order to predict exposures to humans and ecosystems via air, water, and food. Verified models are needed to assess previous conditions and to predict future exposures.

<sup>\*</sup>Principal Investigator, E-mail: joet@lanl.gov

Contaminant transport modeling at the Nevada Test Site (NTS) has been an ongoing activity as part of weapons testing. Now that we are in a testing moratorium, the emphasis has shifted toward examining potential groundwater contamination from the residues of nuclear tests. The Cambric event at the NTS has been the subject of a series of long-term radionuclide migration studies to assess the transport of radioactive species in the subsurface after the detonation of a nuclear device.<sup>1</sup> The low-yield (0.75 kt) Cambric test was conducted below the water table in Frenchman Flat in 1965. From 1975 to 1991 a well installed 91 meters away was pumped in order to draw mobile radionuclides from the detonation cavity. The extracted water was tested for tritium, Kr-85, and other radioactive species. Although both tritium and krypton are conservative tracers in groundwater (i.e., they do not interact with the geologic media through which the water moves), the elution of these two tracers was different at Cambric. The peak of the krypton elution was delayed relative to that of tritium. In addition, while over 92% of the tritium was eluted, only about 43% of the Kr-85 appeared at the pumped well. The causes for these difference were not clear, but the extensive data set offered the opportunity to develop a model that would improve our understanding of contaminant transport in groundwater.

Our immediate objectives were to develop a conceptual framework for explaining our observations at the Cambric site and to apply a numerical model to test the hypotheses. Longer range goals included identifying those factors critical to transport of volatile contaminants in groundwater and evaluation of numerical models suitable for application to contaminant transport problems.

# Importance to LANL's Science and Technology Base and National R & D Needs

Los Alamos National Laboratory (LANL) has state-of-the-art technologies for trace-gas analysis and for environmental transport modeling. The Cambric data set is unique because of its extent and because of the hydrologic setting. Our experience with the TRACR3D code will provide more assurance that the dominant transport processes have been included and that the model can be applied to other sites less well characterized. While the Cambric site is not in itself a health hazard, radioactive noble gases and volatile organic solvents are common contaminants at other Department of Energy facilities. Our analysis of the Cambric data set should help us evaluate the release of volatile materials from groundwater over long times and may provide an important resource for future groundwater transport modeling. 2

#### Scientific Approach and Accomplishments

Our hypothesis was that the carbon dioxide gas that evolved from the carbonate rock around the nuclear device contributed to the post-detonation migration of Kr-85 throughout the rubble chimney above the collapsed crater. This occurred because the working point of the Cambric device coincided with a stratum concentrated in secondary carbonate material; the result was an evolution of about three times the amount of noncondensable gas normally expected in a device of this yield. The tritium vapor condensed in the immediate cavity region and was not affected by the carbon dioxide. As the chimney collapsed, the gas moved upward, becoming distributed throughout the entire length of the chimney. If that chimney reached above the water table, "excess" CO<sub>2</sub> would have been released into the vadose zone. The behavior of this gas is particularly important to consider, since Kr-85 is a gaseous radionuclide; hence, where CO<sub>2</sub> moves, so does Kr-85.

After chimney collapse, the gases were distributed throughout the chimney, with some "lost" above the water table. With the Kr-85 distribution different from the tritium distribution immediately in and around the cavity, we can understand the late elution of krypton relative to tritium as well as the loss of over half of the krypton. We then began to apply numerical models of increasing complexity to evaluate this scenario. A variety of numerical simulations were performed using the finite difference code TRACR3D. Analyses were conducted with a wide range of assumptions since there is uncertainty about some of the Cambric field conditions. Using a calibration method based on the tritium elution curve, we obtained numerical simulations consistent with the suggested difference in the krypton and tritium distributions prior to pumping. However, beyond the argument of transport with carbon dioxide, we found additional factors that may have affected the initial radionuclide distribution at the Cambric site. One issue that does not appear to have been previously explored is that of thermally induced convection currents. We spent some time exploring the possibility of buoyancy driven flow and how it may have affected the distribution of radionuclides prior to satellite well pumping. A second issue regards whether shallow natural hydraulic gradients may have caused migration of nuclides during the 10 years before pumping of the satellite well.

Despite the extensive data from the field study, there are still uncertainties in some of the hydrologic parameters, making it impossible to conclusively resolve the results at Cambric. This study is not the final word on Cambric but does advance our ability to model a complex system and to identify significant parameters in contaminant transport.

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## References

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[1] Thompson, J., "Radionuclide Migration Studies at the Nevada Test Site," Radiochim. Acta 54 149-154 (1991).